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13. ABSTRACT *(Maximum 200 words)*
This report describes the purchase specifications and initial use of a dual gated imaging system capable of intensified imaging with gate times a short as 1 nsec. The system consists of two primary subsystems: (1) dual fast gated image intensifiers, and (2) a sensitive CCD camera and readout/computer system. With dual images, it is possible to measure the fluorescent lifetimes of the dopant which is the marker for the fuel, fluoranthene, in a two dimensional image, and subsequently to correct the fuel intensity image for oxygen quenching. From the fuel and oxygen images, the equivalence ratio image can be calculated. In initial experiments, the system has been used to capture instantaneous (<50 nsec) equivalence ratio images for gaseous methane jets and for diffusion flames.

14. SUBJECT TERMS
fluorescence, diagnostics, imaging, equivalence ratio

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Dual Gated Imaging System
for
Enhanced Fluorescent Diagnostics Measurements

Final Report

Lynn A. Melton

March 13, 1992

U. S. Army Research Office

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I. STATEMENT OF THE PROBLEM STUDIED

Recent work at the University of Texas at Dallas has demonstrated that (1) both fluorescence intensity and lifetime information can be obtained from images captured on array detectors and (2) this information can be further processed to yield images of the fuel/oxygen equivalence ratio, the key mixing parameter in combustion studies. The imaging of the lifetimes requires two sequential images taken only a few nanoseconds apart, which in turn requires the use of two fast, gated two-dimensional detectors. The purchase of such a Dual Gated Imaging System was funded through this Army Research Office grant. This report describes the components purchased and the initial equivalence ratio imaging results obtained for a gaseous methane jet into air and for a methane diffusion flame. The ability to capture an image of the instantaneous equivalence ratio is important and can assist in the understanding of many combustion problems.

Recently, "proof of concept" experiments have been published which show how sequential images of the oxygen-quenched fluorescence of fluoranthene could be used to display two-dimensional, instantaneous, non-intrusive images of the fuel/oxygen equivalence ratio.[1] Those "proof of concept" experiments were carried out with a single, fast-gated, one-dimensional diode array detector; the experiments showed, however, that with two fast-gated, two-dimensional array detectors it should be possible to obtain instantaneous images of the fuel/oxygen equivalence ratio.

Two key ideas make the imaging of the fluorescent lifetimes feasible. First, Ballew and Demas have recently published an algorithm for rapid lifetime determination which allows numerically-stable calculation of lifetimes from only two time-integrated intensity measurements, i.e., only two images are required.[2] Second, fluoranthene has been found to be an unusual and appropriate fluorescent dopant; its fluorescence is quenched by molecular oxygen at only 1% of the gas kinetic collision rate.[3] Thus fluoranthene in 0-4 atmospheres of oxygen provides a range of fluorescence lifetimes in the 10-30 nsec range.

A common and important criticism of diagnostic experiments based on fluorescent dopants is that quenching of the fluorescence, particularly by the oxygen in combustion systems, causes substantial uncertainty in the interpretation of the results: a low intensity from some region of the image may result from a low vapor concentration <u>or</u> from a high vapor concentration and a high oxygen concentration. However, the use of dual, fast-gated images can turn the oxygen quenching effects to an advantage. Both the fluorescence intensity and the fluorescence lifetime depend upon the oxygen concentration.

With the dual gated imaging methods, the use of two sequential gated images, separated by a few nanoseconds, allows the calculation of the fluorescence lifetimes. With the addition of laboratory measurements of unquenched lifetimes and oxygen quenching rates, the fluorescence lifetimes may be used to calculate the local oxygen concentrations. These, in turn, can be used to correct the intensity measurements for the effects of oxygen quenching. Thus, one obtains images of (1) the fuel concentration, (2) the oxygen concentration, and (3) from pixel-by-pixel ratios, the fuel/oxygen equivalence ratio.

II. SUMMARY OF MOST IMPORTANT RESULTS

A. Purchase of Dual Gated Imaging System

1. Rationale

The original budget described a system for dual gated imaging which involved the purchase of separate components such as image intensifier tubes, high voltage pulsers, bias power supplies, etc., the construction of impedance transformers, and the assembly of these components into a system. In the process of contacting suppliers of these components, it was found that Grant Applied Physics could supply a dual gated optical imaging system. With this commercial system, it would be possible to get the imaging experiments up and running much quicker since the technological problems associated with integration of the nanosecond, high voltage pulses would have been solved by Grant Applied Physics. However, the Grant Applied Physics dual gated optical imaging system cost $62,000, and thus in order to meet the constraints of the budget for this grant, only one Photometrics STAR-1 CCD camera system was purchased. The output images from the two intensifiers were then focused onto the single CCD detector, a strategy which allows the imaging experiments to be carried out with a loss of a factor of two in spatial resolution.

An important consideration in the decision to purchase the system described above is that it can be upgraded, if desired, without significant technical problems. The current experiments can be carried out by focusing both images onto a single CCD detector, but if later, more sophisticated, experiments require it, a second CCD camera could be added (approximately $17,000). The Grant Applied Physics pulser, which currently provides gate pulses of 1, 2, 4, 8, or 12 nsec, is ideal for the capture of lifetime images down to 1 nsec, but, if desired, the pulser could be modified to provide additional gate pulses of 0.25 and 0.50 nsec (approximately $10,000).

2. Equipment

a. CCD Camera

Photometrics STAR-1 CCD camera system, with controllers and memory for single image, cooler/recirculator, GPIB interface, Thompson 7883 CCD chip (384 x 576 pixels) with Metachrome II (UV enhanced) coating, and software for computer control of CCD controller

$17,825

b. Image Intensifiers, Pulsers, and Power Supplies

Grant Applied Physics two channel gated optical imager for UV use, lens coupled, fast gate times of 1,2,4,8, and 12 nsec, slow gate times (300 microsec), and DC, multichannel plate intensifiers, 18 mm diameter cathode (minimum gate time <120 ps)

$62,000

B. Initial Equivalence Ratio Imaging Experiments

The gated image intensifier system was delivered in late November 1991. In February 1992 successful equivalence ratio imaging experiments were carried out. Results were obtained for both a gaseous methane jet into air and for an ignited gaseous methane jet into air (i.e., a diffusion flame). The ability to obtain instantaneous equivalence ratio images for a diffusion flame is illustrative of the enormous potential of these dual gated imaging methods.

Figure 1 is a schematic diagram of the dual gated imaging apparatus. Figure 2 shows how the relative involatile fluoranthene (NBP 375 ^{o}C) is seeded into the methane flow. The temperature of the chamber was maintained at 169 ^{o}C, which provides a vapor phase concentration of fluoranthene sufficient to produce fluorescence intensity approximately equal to that of 1×10^{-6} M fluoranthene in cyclohexane. The tube diameter is 9.5 mm.

The imaging experiment was carried out slightly differently from the experiment proposed earlier.[1] The gate width for one intensifier was set to 20 microseconds in order to be sure of capturing all of the "fuel image" fluorescence; in this manner it was not necessary to assume that the laser pulse width (8 nsec FWHM) was short compared to the fluorescence lifetimes to be measured (20-30 nsec). The second pulse (12 nsec width) was delayed 55 nsec from the rising edge of the laser pulse. The ration of the intensities obtained from these two gate pulses was calibrated against known oxygen pressures, and this calibration curve was used to interpret the gaseous jet/diffusion flame data. Spatial intensity variations within the laser sheet were determined by irradiation of a cuvet containing 1 x 10^{-6} M fluoranthene in cyclohexane; raw images were normalized to remove these spatial variations.

Figures 3-6 show the results obtained. Each image is 92 pixels wide x 155 pixels high; the magnification corresponds to 32 pixels/inch of actual dimension. In each set, there is an image of (a) the methane pressure, (b) the air pressure, and (c) the equivalence ratio. Each set of images (a-c) was obtained from a single laser pulse and thus are simultaneous information about the jet/flame. The air pressure image was obtained by analysis of fluorescence lifetime data, i.e., from oxygen quenching. In regions where the fluoranthene fluorescence intensity was too low to obtain lifetime data, it was assumed that the air pressure was one atmosphere. The methane image was obtained by using the air pressure image and known quenching rates to correct the methane image data for oxygen quenching. In order to obtain absolute scaling of the methane image, it was assumed that the methane pressure at the nozzle exit was one atmosphere. Note therefore that the methane and air images are somewhat independent; it was not assumed that the sum of the methane and air pressures equalled one atmosphere except at the nozzle exit and in regions where the measured fluorescence intensity was minimal.

Analysis of the diffusion flame data, figures 4 and 6, requires the assumption that the photophysics and photochemistry of fluoranthene, particularly the oxygen quenching rate, is independent of temperature. This assumption has not been tested, and thus attempts at quantitative interpretation of these images should be tempered.

In figure 3c, the displayed equivalence ratio distribution is substantially broader than that for methane or air. This is the result of the choice of a logarithmic gray scale for the equivalence ratio as opposed to the linear gray scale for the methane and air pressures. Nonetheless, it is interesting to note in figures 3c and 4c the existence of pockets of well mixed fuel substantially above the primary jet/flame. These are not due to noise in the measuring system. Figures 5 and 6 show the dramatic effect of flame buoyancy on the gas distribution. Figure 6c, particularly, shows that, even with a logarithmic display of equivalence ratio, there is a well defined flame front with very little air inside the flame front and very little fuel outside the flame front.

C. Conclusions

The dual gated imaging system was ordered, and although delays in the delivery of the gated intensifiers were frustrating, the system was put into good working order quickly. The images obtained so far show that the system is now capable of use in flow/mixing/combustion experiments. Further applications will follow under alternate ARO funding, particularly grant DAAL03-91-G-0033.

III. LIST OF PUBLICATIONS AND TECHNICAL REPORTS

No publications or technical reports have resulted. An initial paper describing the successful equivalence ratio imaging experiments is in preparation.

IV. PARTICIPATING SCIENTIFIC PERSONNEL

No personnel were supported by this grant. The following individuals worked on the purchase of the equipment and the development of the initial experiments.

1. Lynn A. Melton, principal investigator
   Professor of Chemistry

2. Tuqiang Ni
   Postdoctoral Research Scientist

V. REPORT OF INVENTIONS

None

VI. REFERENCES

1. T.Q. Ni and L.A. Melton, "Fluorescence Lifetime Imaging: An Approach for Fuel Equivalence Ratio Imaging", Applied Spectroscopy, **45**, 938 (1991).

2. R.M. Ballew and J.N. Demas, "An Error Analysis of the Rapid Lifetime Determination Method for the Evaluation of Single Exponential Decays", Anal. Chem., **61**, 30 (1989).

3. L.J. Jandris, R.K. Force, and S.C. Yang, Applied Spectroscopy, **39**, 266 (1985).

VII. List of Figures

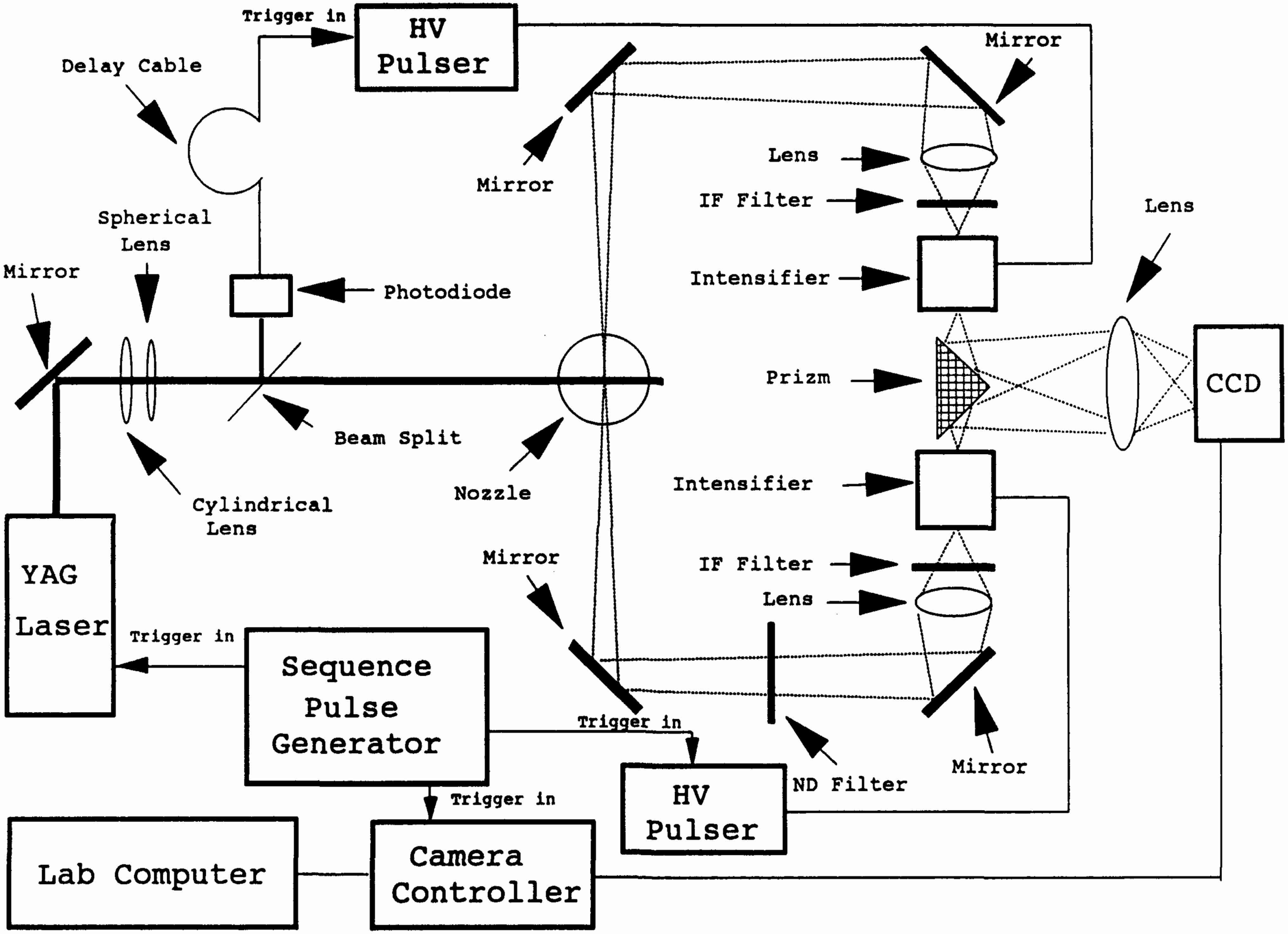
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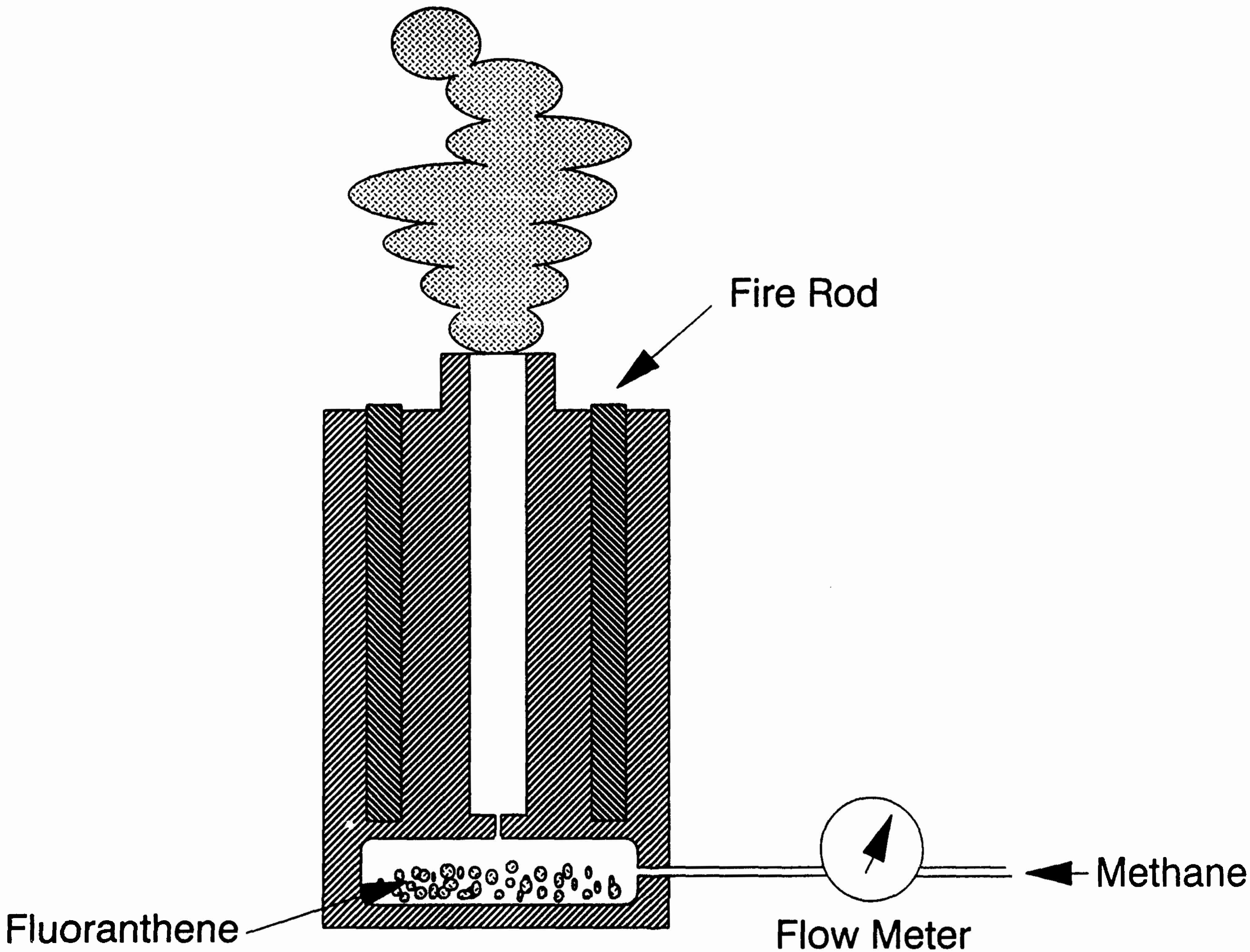
Figure 1



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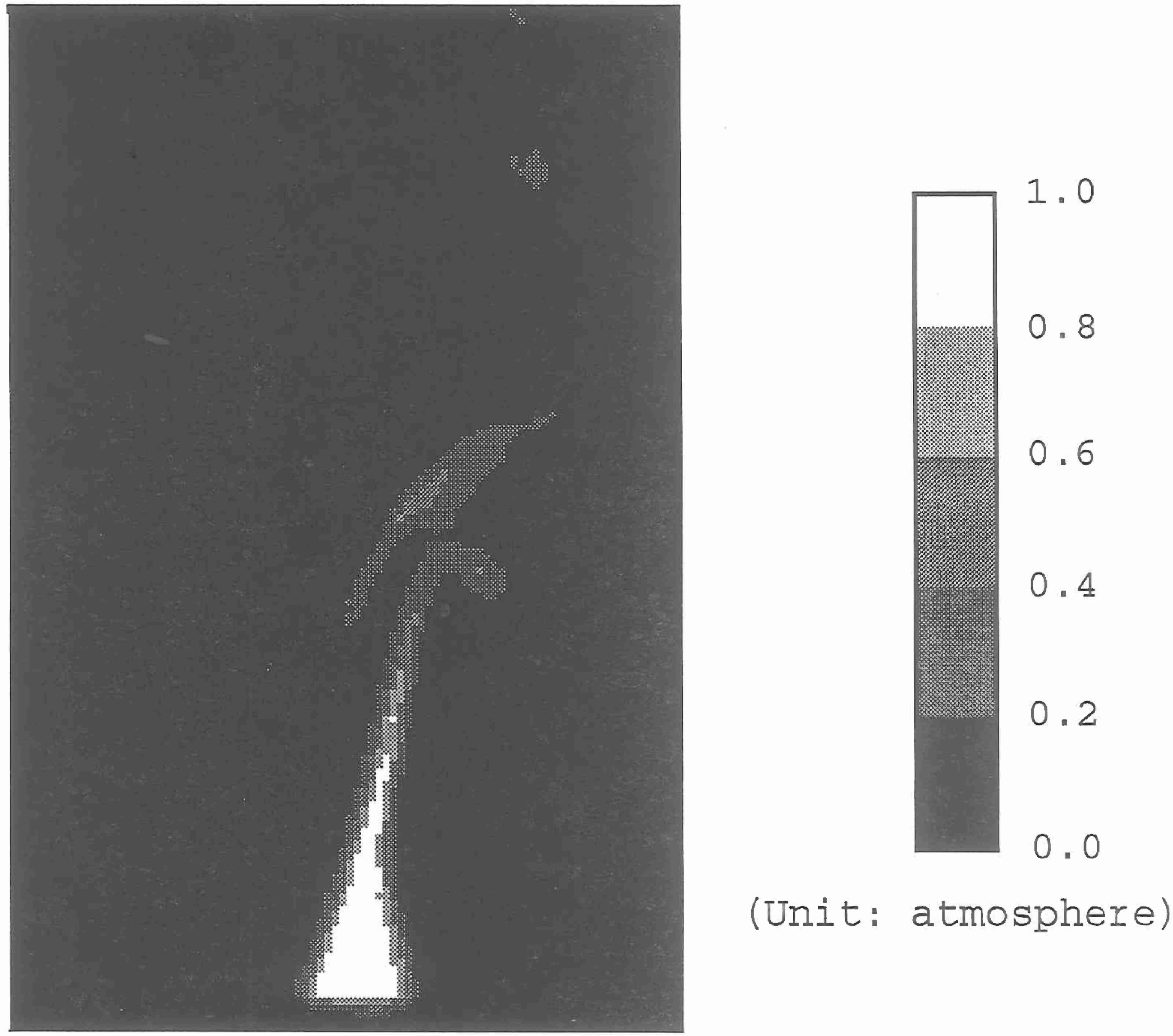

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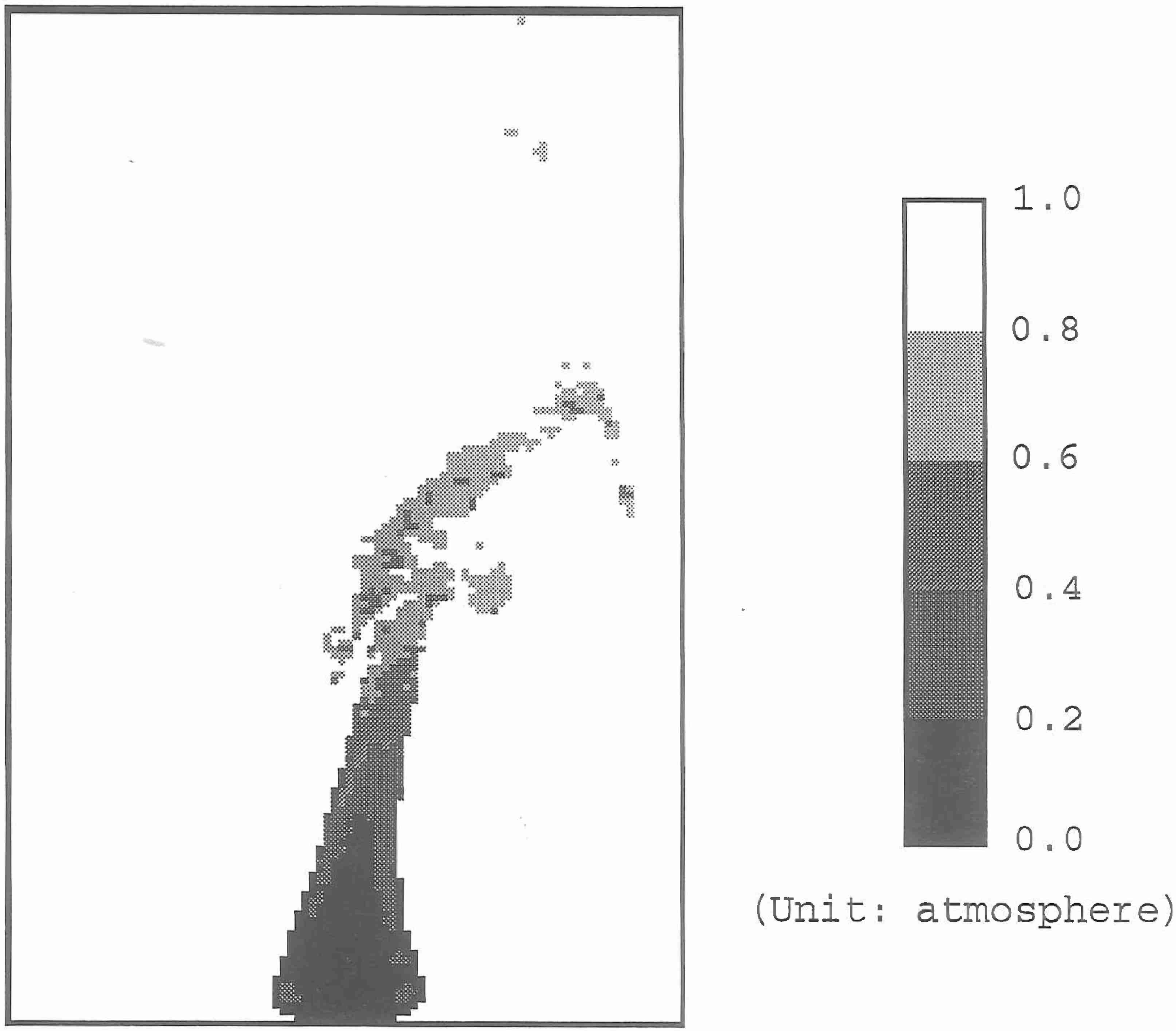

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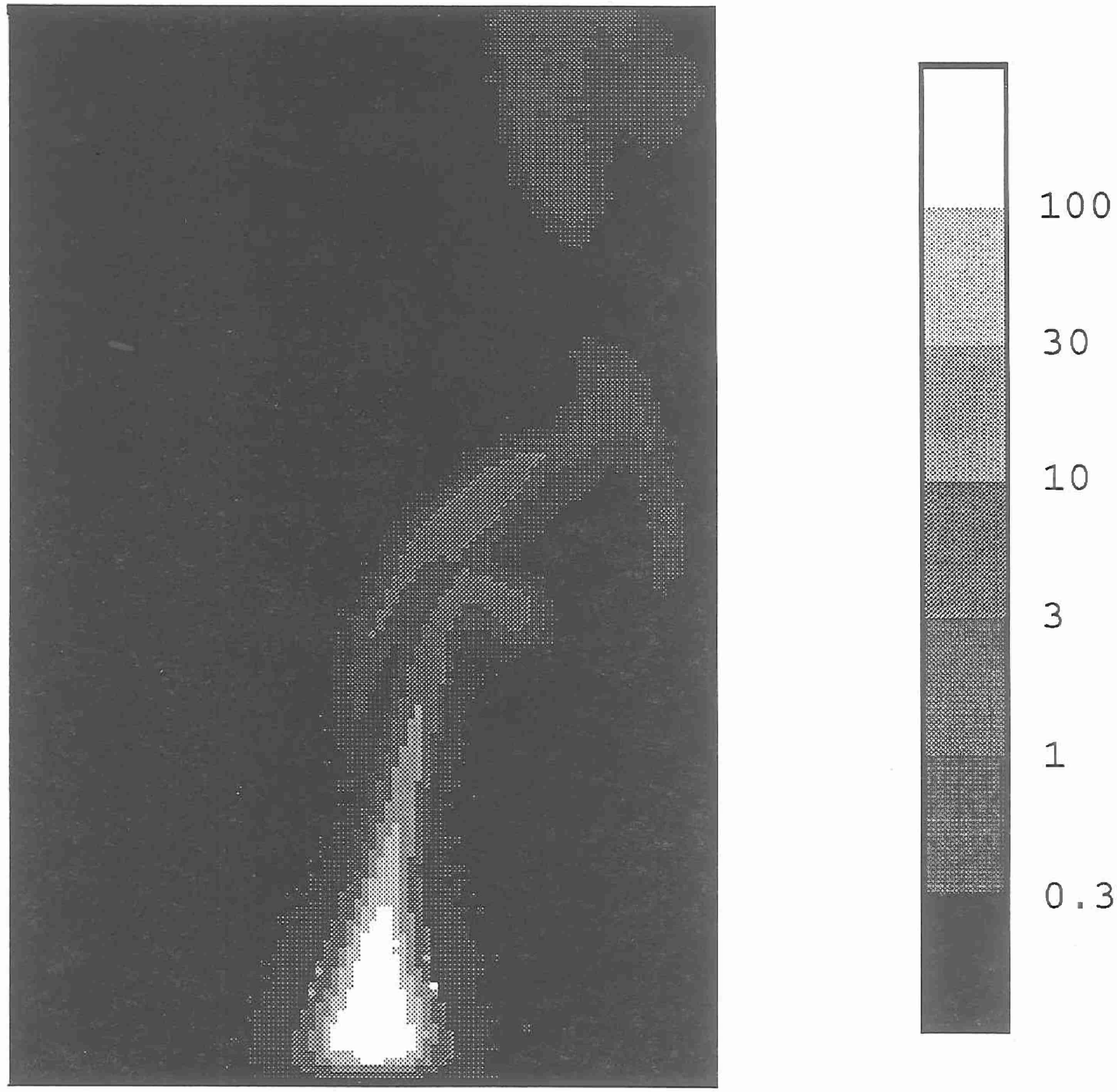

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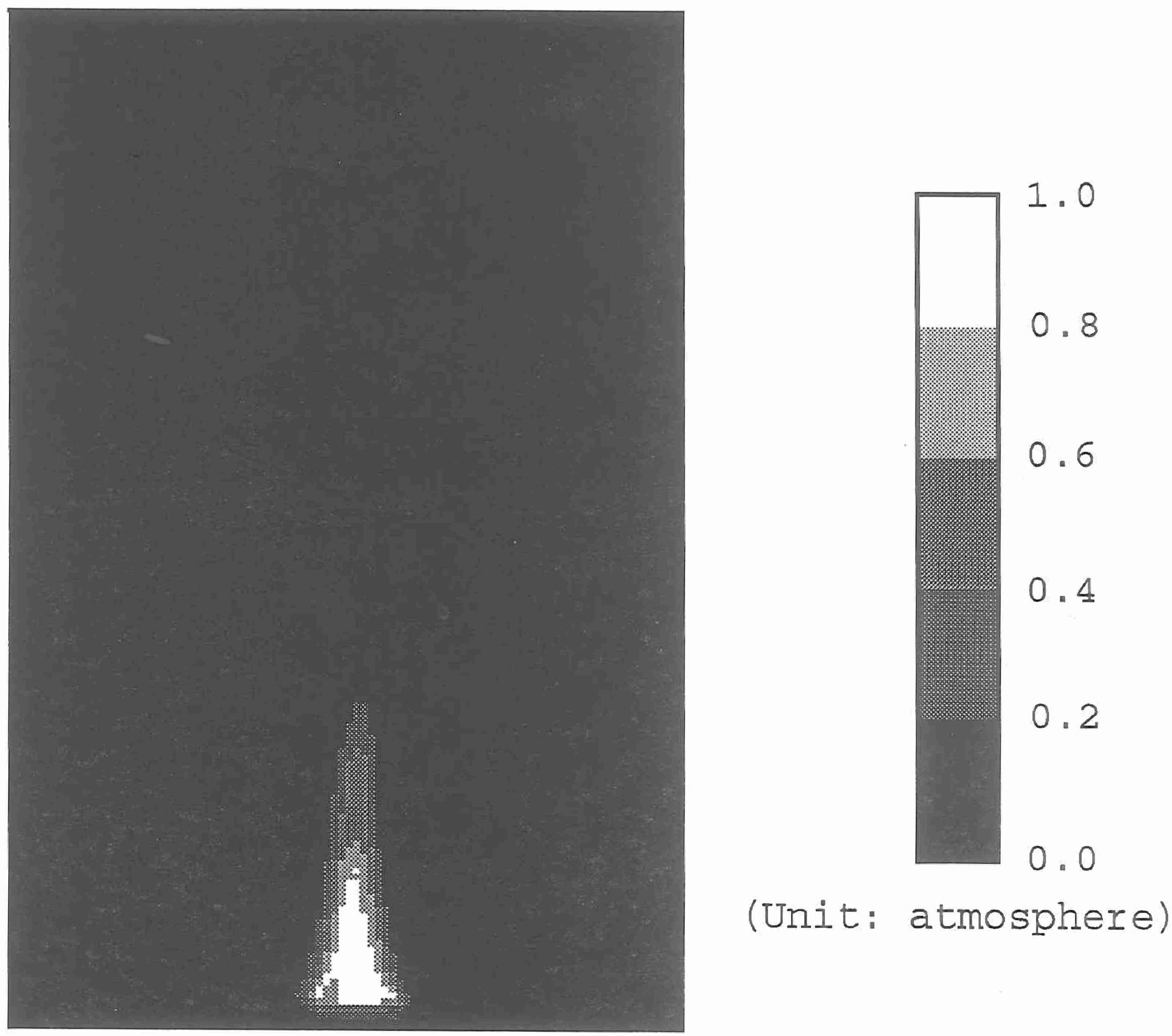

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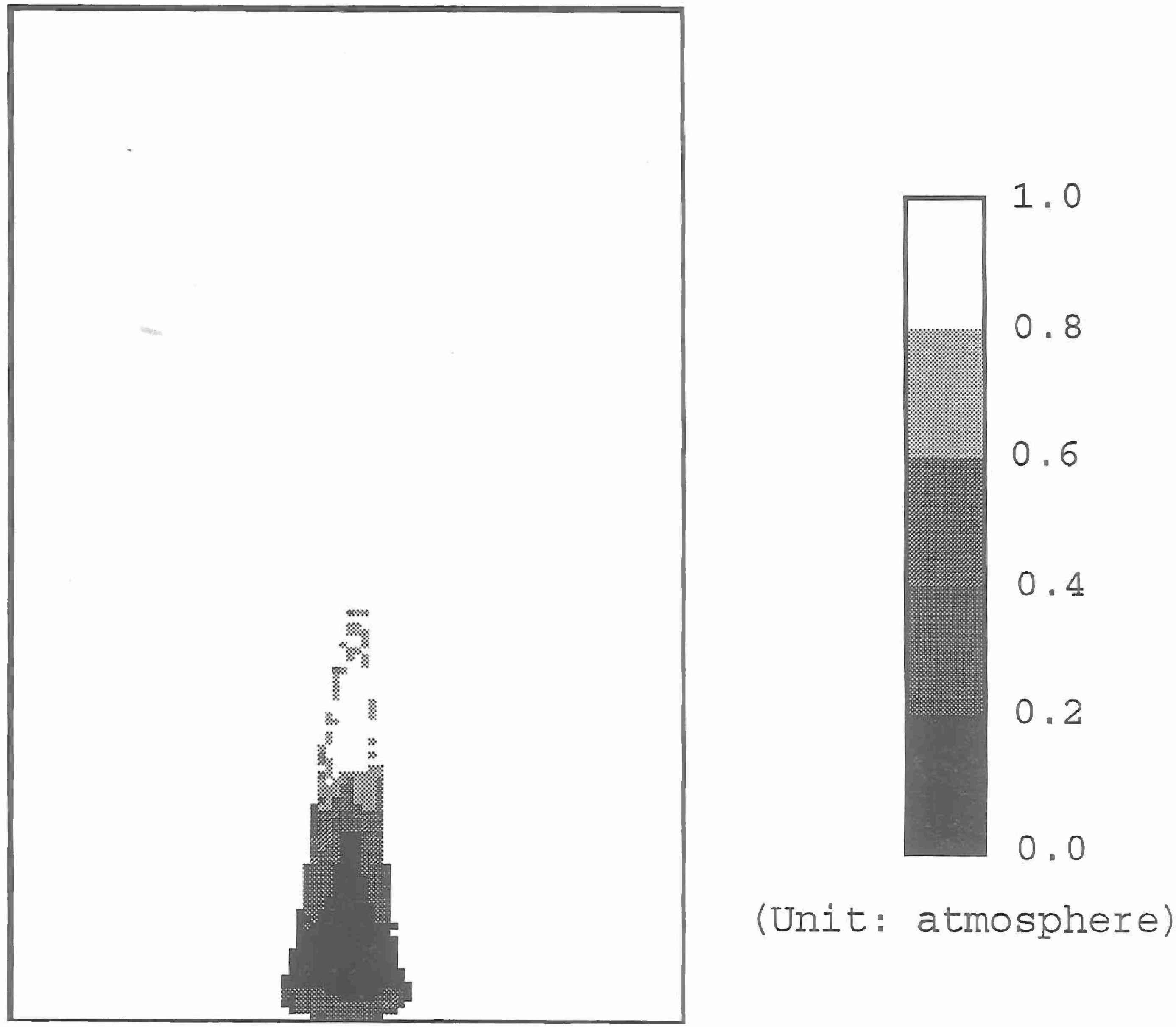

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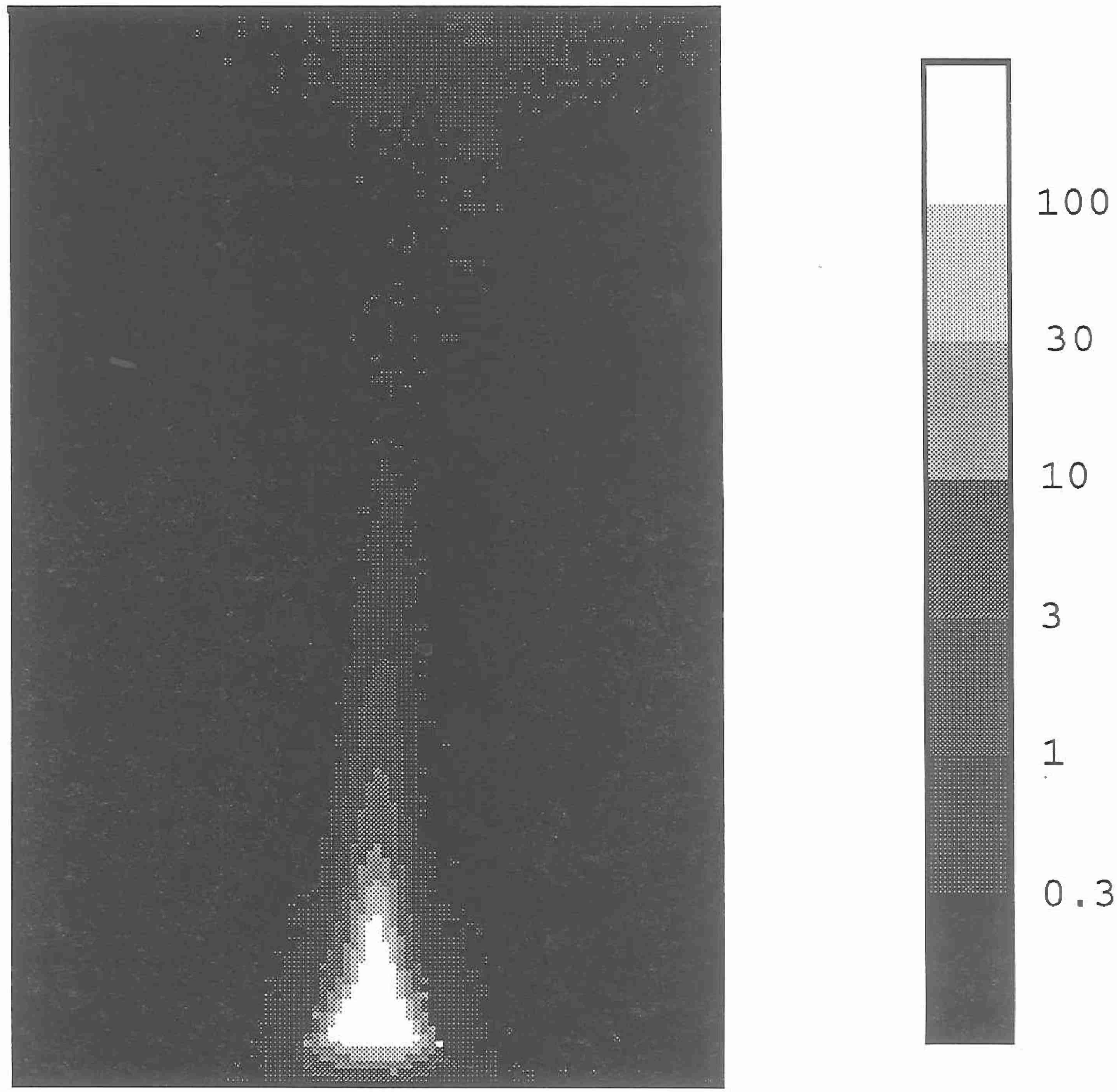

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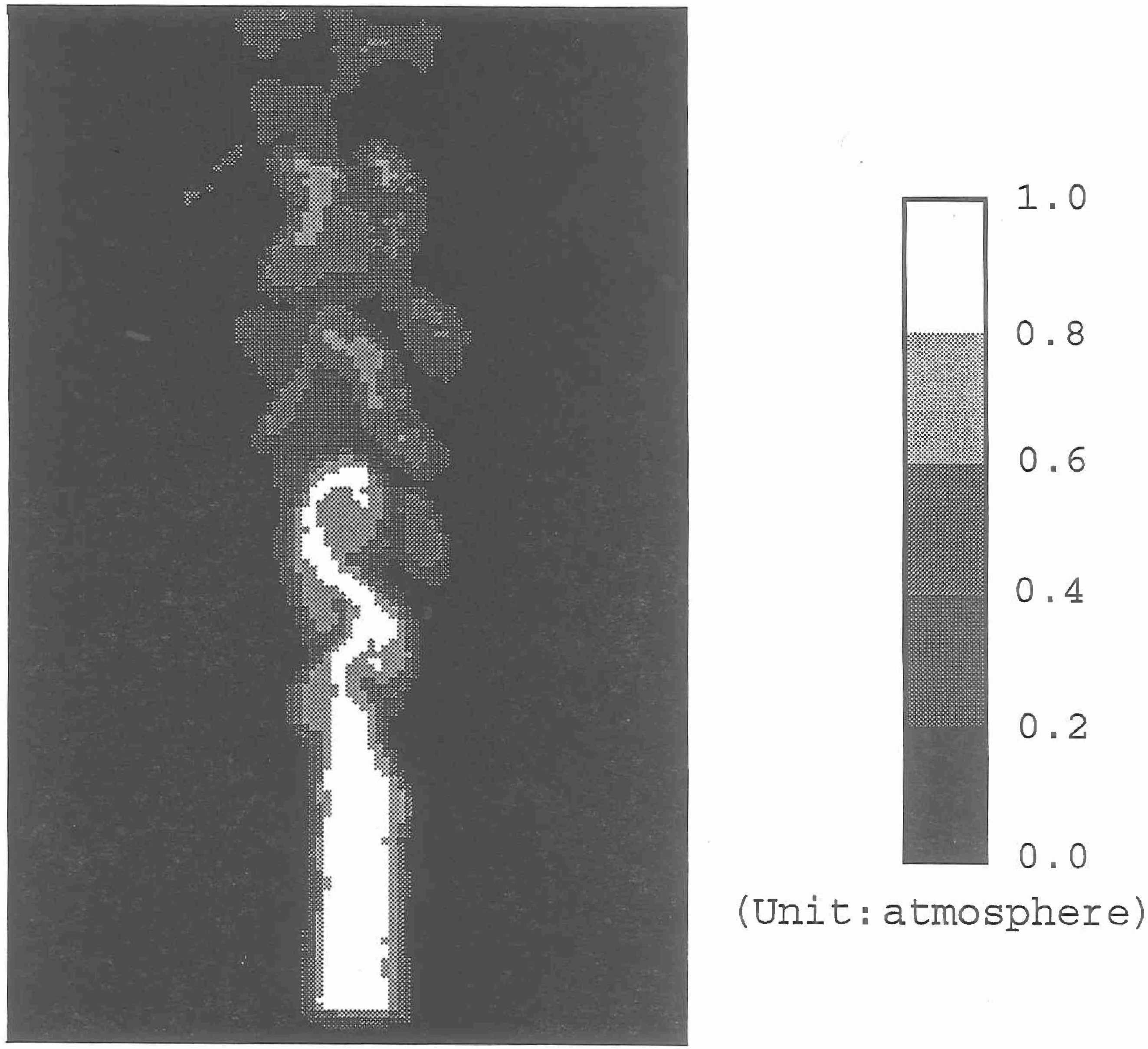

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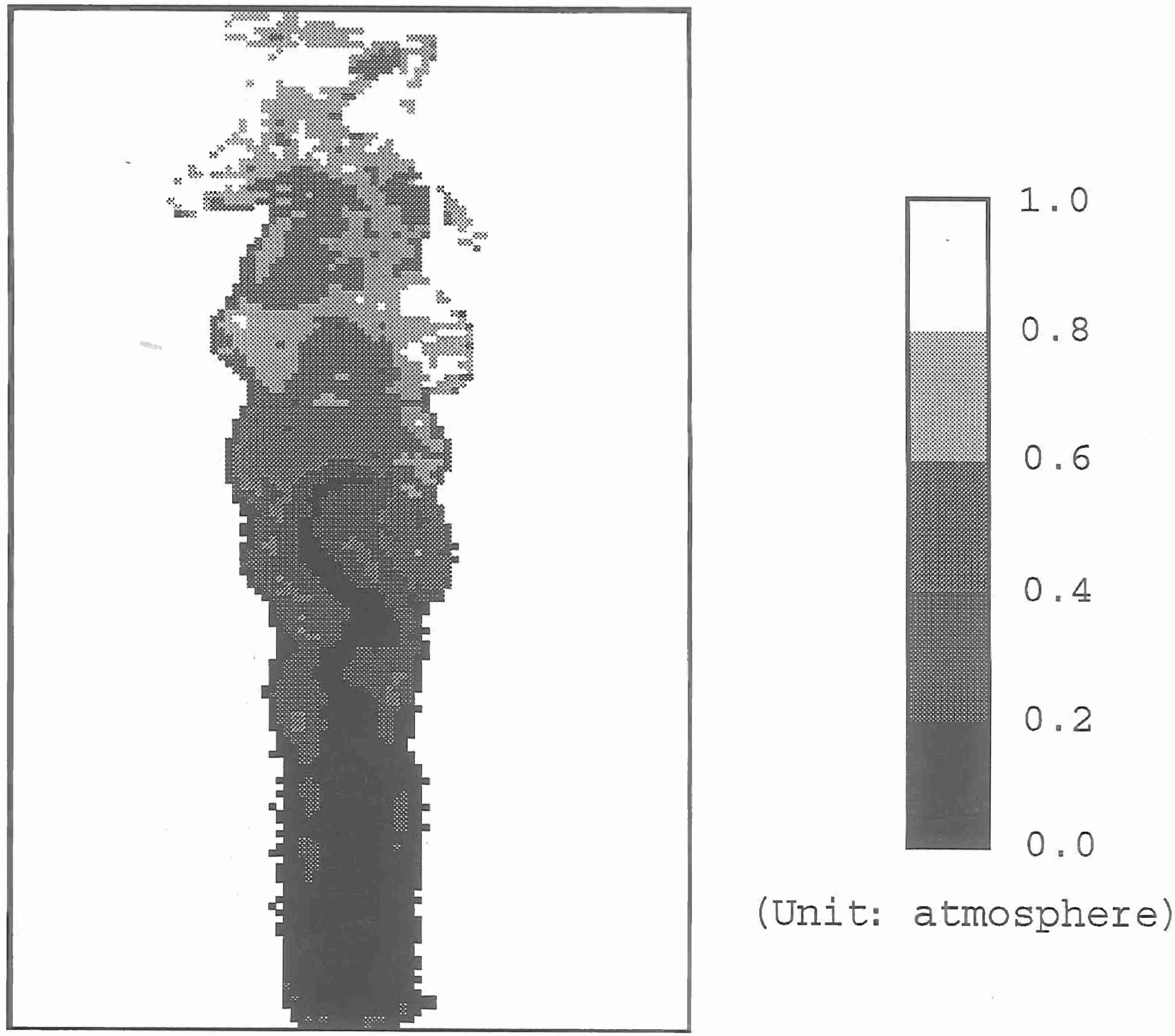

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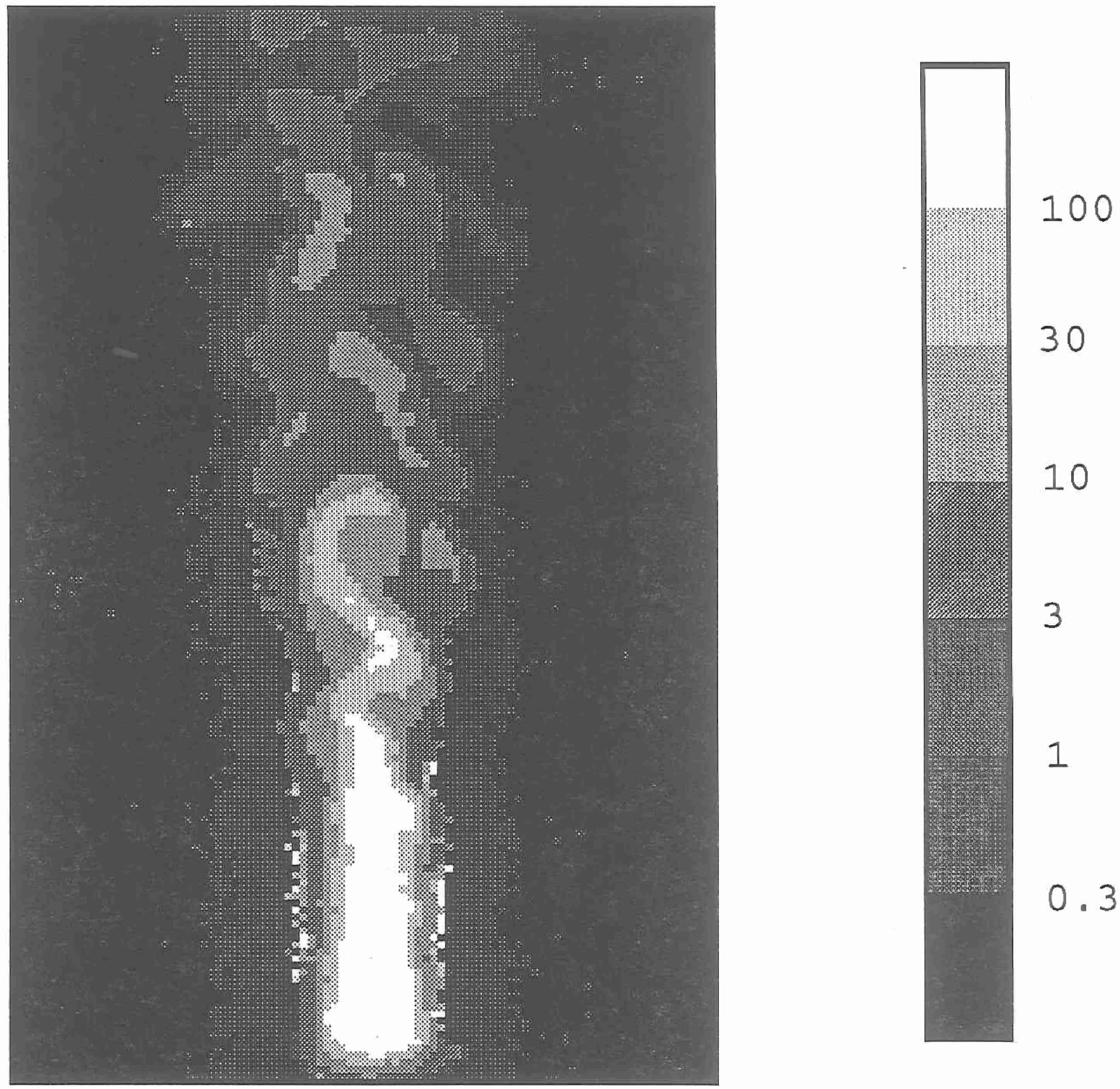

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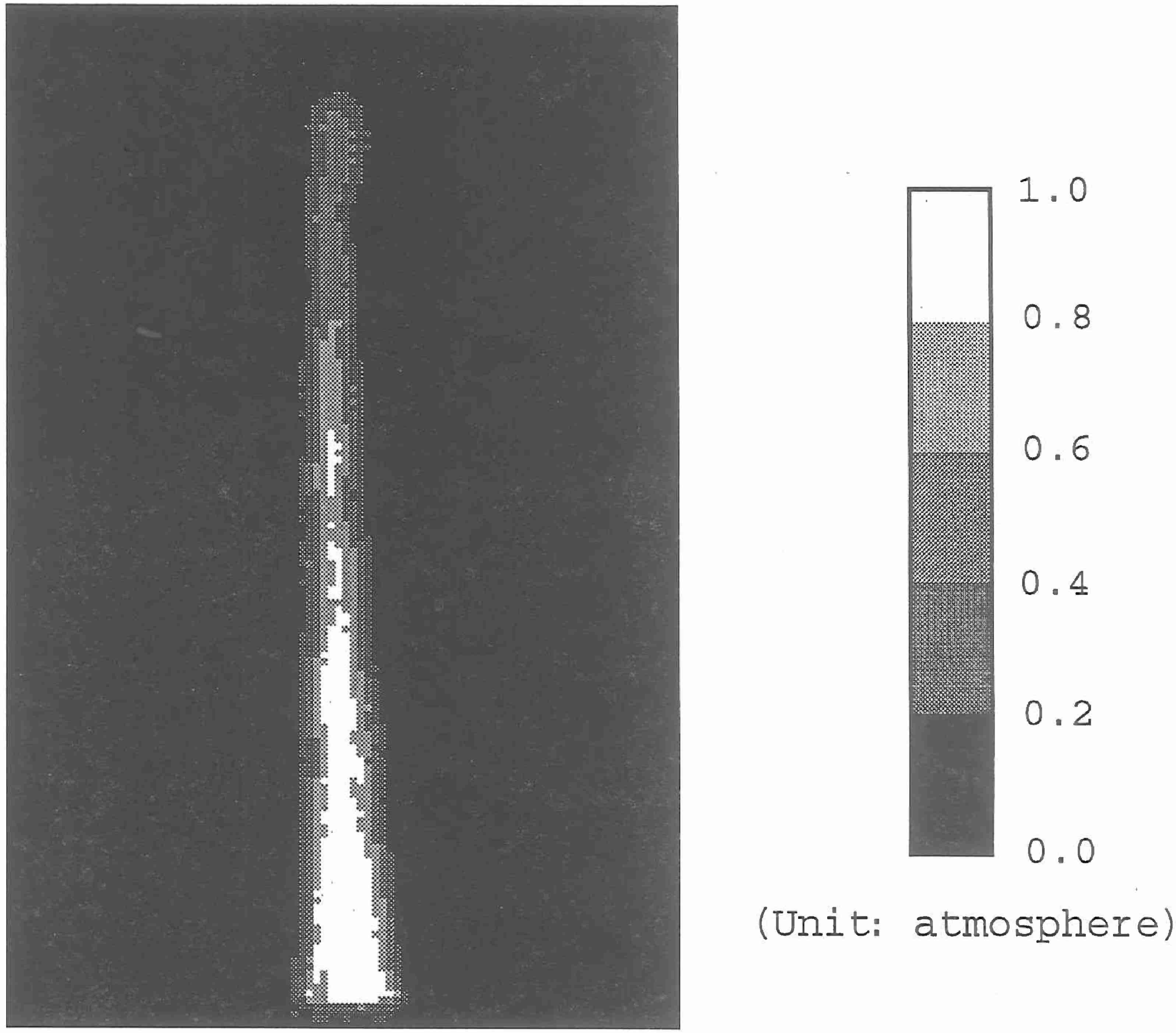

Figure 6a

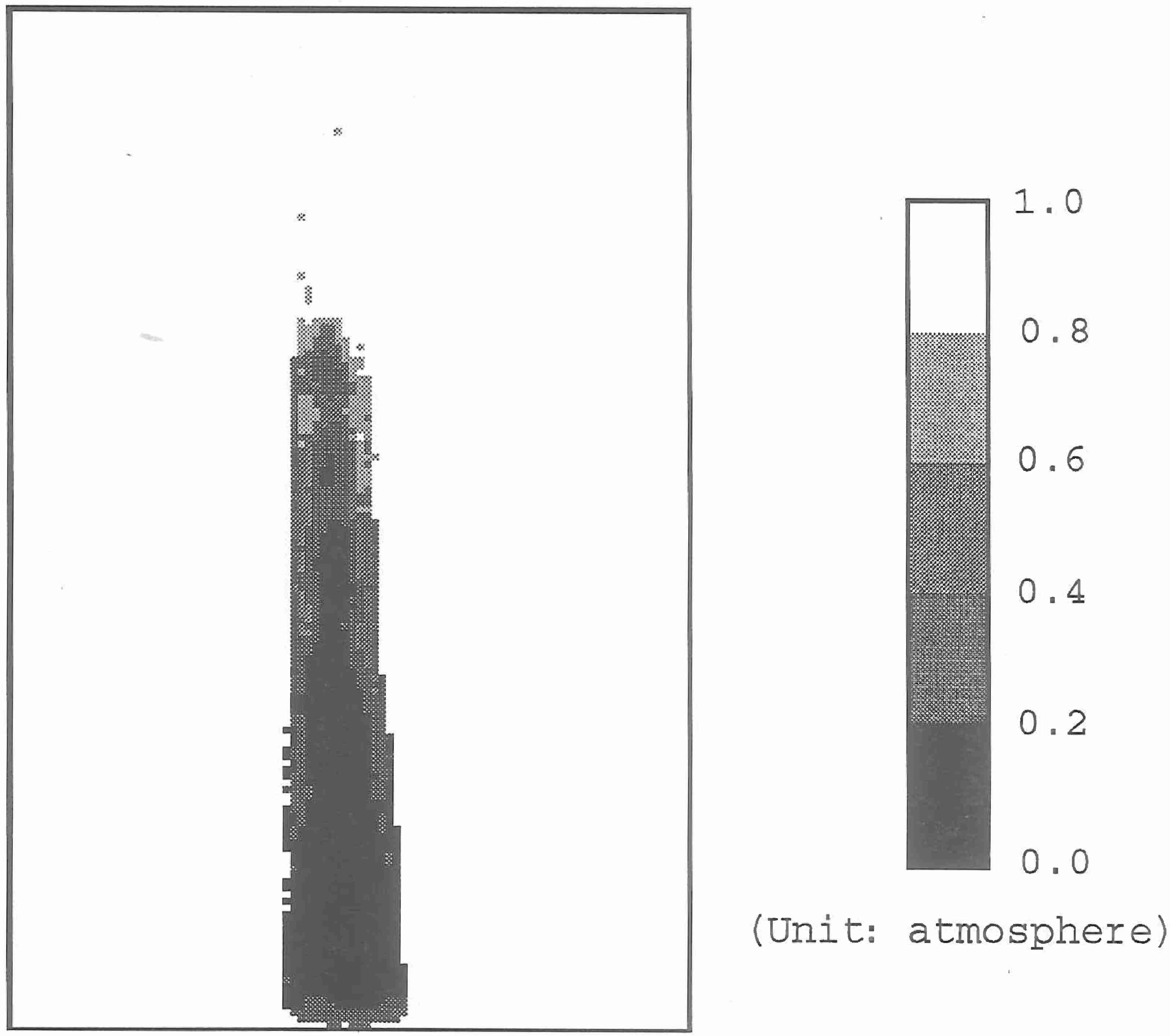

Figure 6b

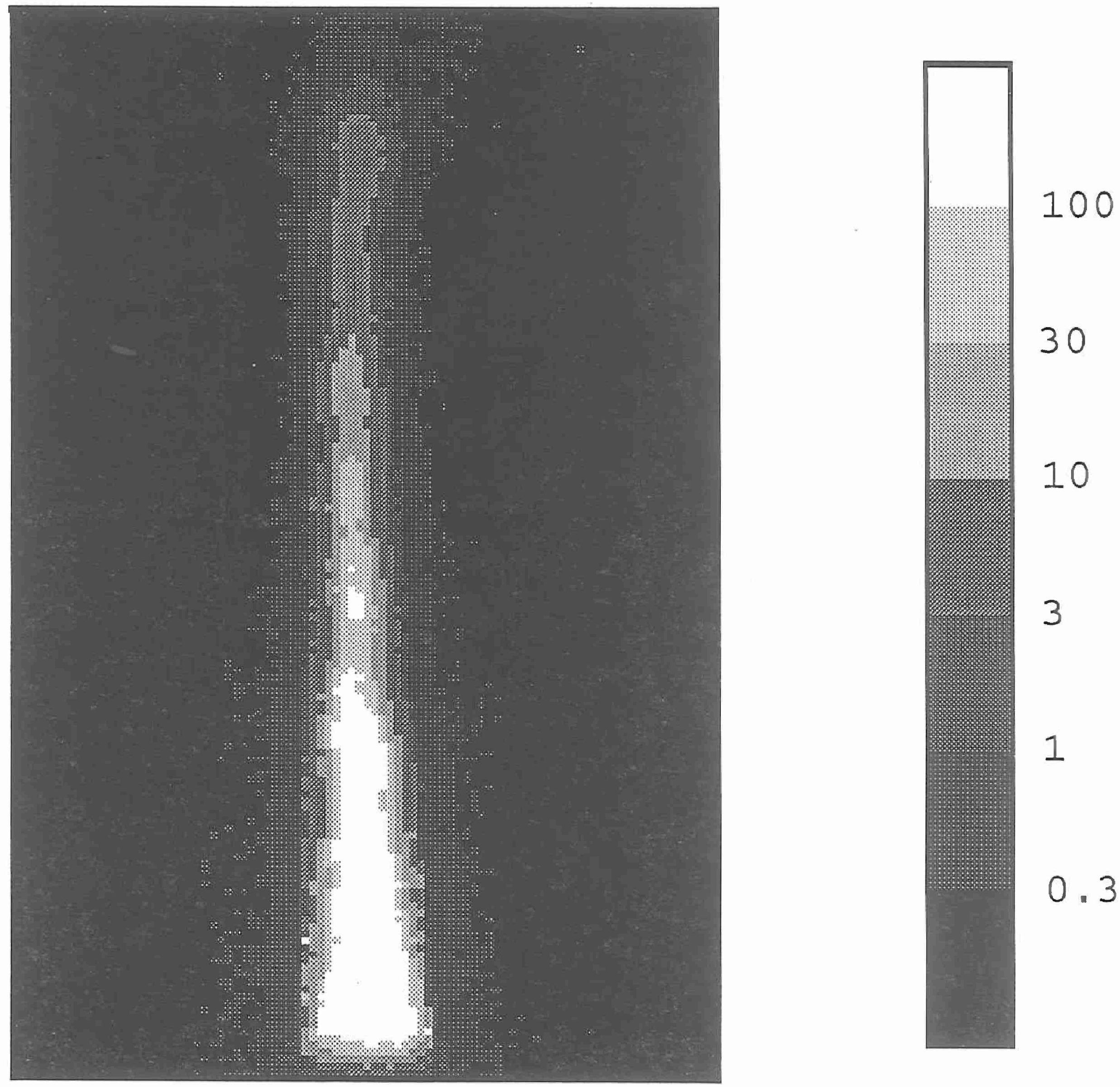

Figure 6c